Ultrashort Laser Pulse Propagation in Water

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LONG-TERM GOALS

The major objectives of this proposal are twofold. The first objective is to perform both an experimental and theoretical study of the factors affecting the propagation distance behavior of ultrashort (femtosecond) laser pulses in water. This study will be conducted in the so-called linear regime which involves laser intensities that are below the threshold where nonlinear effects set in. A fundamental problem, which will be resolved by this research, is whether or not the temporal width and spacing of short pulses affects its absorption spectrum. If one simply knows the spectral composition of the input pulse and the absorption spectrum of the water, is this sufficient to predict the temporal evolution of the pulse? If such is not the case, then we will have to consider the way the system responds to pulse widths and pulse spacing which are short compared to vibrational relaxation times in water.

The second phase of the research will be to explore the nonlinear regime where dramatic changes to the temporal, spatial, and spectral properties of the medium occur. The primary processes being self-focusing and self-phase modulation due to the Kerr effect (also called the quadratic electro-optic effect which was discovered by John Kerr in 1875). Self-focusing can lead to an enormous increase in the peak intensity where long filaments can occur and in some cases lead to supercontinuum generation or "white light" generation first discovered in 1970.

OBJECTIVES

The propagation of an ultrashort pulse of light through a linear and absorptive medium such as water, is of great fundamental importance for several reasons. One of the most important of which is that it may be possible to transmit information over much greater distances using ultrashort pulses compared to propagation distances achieved by using pulses with long time durations, including CW (continuous waves). The first prediction of optical precursors was given by both Sommerfeld and Brillouin[1] in 1914 using an asymptotic method now called steepest descent. Their analysis was based on a step-modulated field propagating through a Lorentz dielectric which is nothing more than a collection of damped harmonic oscillators. Later refinements to their conclusions were made by Oughstun and Sherman[2]. The first measurements which claimed to observe optical precursors in deionized water were made by Choi and Österberg[3] where they found that the precursors were attenuated non-

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exponentially with distance. What was extremely significant about this work was that they found the pulse energy detected at a distance of 5 m in water with a 60 fs pulse was one hundred times greater than the signal obtained using a 900 fs pulse (having the same fundamental wavelength) which had the usual Lambert-Beer behavior (It should be noted that the so called Lambert-Beer law was first discovered by Pierre Bouguer in his published work in 1729 titled *Essai d'optique sur la gradation de la lumiére* and we will hereafter refer to it as the Bouguer-Lambert-Beer Law or BLB. The law is valid for monochromatic sources but unfortunately researchers have mistakenly applied it to pulse propagation where the bandwidth covers changes in the absorption coefficient). Their conclusions were brought into serious doubt by Alfano[4] et al. who claimed that their bandwidth was not wide enough to cross the 760 nm absorption band of water and therefore that their conclusions were questionable.. In a later paper, Fox and Österberg[5] found deviation from BLB when they used pulses with temporal widths of 60 fs and a repetition rate of 1 kHz where they observed more than two orders of magnitude less absorption after propagation through 6 m of water when compared to BLB. They also found that BLB was not violated for pulses with varying bandwidth with temporal widths of 900 fs (pulses centered at 800 nm) and repetition rates of 80 MHz. Li et al[6] have performed a more recent measurement for liquid water and they found deviations from the BLB law after the pulse had propagated only 1.5 m and the deviation continued to increase out to 3.5 m which was as far as they could detect the signal. The shortest pulse they used was 10 fs with a repetition rate of 75 MHz which gives a pulse spacing of 13.3 ns. They showed that deviation from BLB behavior only occurred when the pulse bandwidth was of the order of 100 nm but for shorter bandwidths (< 30 nm) BLB behavior was verified. It is important to note that this pulse spacing is much longer than the vibrational relaxation times in liquid water which are typically of the order of a picosecond. The most recent experiments by Okawachi et al.[7] used four different laser systems centered at 800 and 1530 nm to show the dependence of pulse duration and repetition rate on pulse absorption. Their bandwidths ranged from 20-60 nm and they showed that BLB behavior was obeyed in every case. It is now clear that there is a real enigma associated with the entire phenomena of ultrashort laser propagation in water.

APPROACH

Experimental:

There are three basic improvements over what has been done in the past that we are proposing:

(a) Improved input pulse shapes

The laser system that we will use in this experiment is the one we currently use for femtosecond CARS spectroscopy of dipicolinic acid and bacterial endospores[8-11]. We have an amplified femtosecond laser (Mira + Legend, Coherent) whereby we obtain up to 1 mJ per 35 fs pulse (at 1 kHz repetition rate) at the fundamental 800 nm wavelength. We can control the chirp (positive or negative) of these 800 nm pulses over a wide range (increasing the pulse duration by up to two orders of magnitude, to over 3 ps, while keeping the spectrum unchanged) very easily, and very precisely by adjusting our pulse compressor. Thus we will be able to study propagation of laser pulses of different length but having identical spectra. Fig. 1 shows the experimentally measured pulse shapes obtained by this method and measured by our home-made variation of frequency-resolved optical gating (FROG).

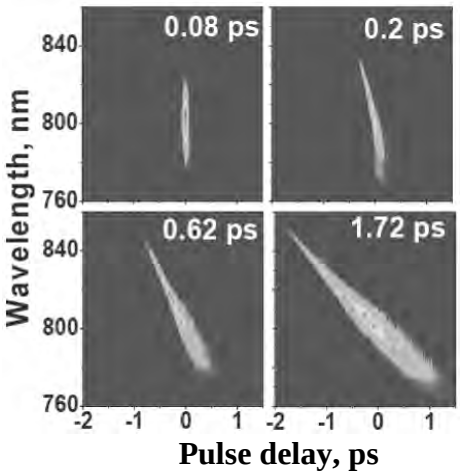

Fig. 1. Chirped pulses obtained by adjusting the pulse compressor and measured by a modified frequency-resolved optical gating technique. The numbers in the upper right corner of each plot give the pulse duration; the spectra of all four pulses are identical.

Alternatively, we can modify the pulse spectral intensities while keeping the spectral phases fixed. Our home-made pulse shaper allows us to make the spectral width up to 100 times narrower and the pulse length correspondingly longer without introducing any chirp. Moreover, our pulse shaper allows us to fine-tune the precise frequency of these narrow-band pulses. We will use such spectrally-modified pulses for the detailed study of light propagation through water. In addition, we have the capability to produce some more complex pulse shapes, should it become necessary or interesting. Our recent paper[12] gives a colorful illustration of potential possibilities, and shows an example of the use of pulse shaping for coherent Raman spectroscopy. More complex pulse shapes will be particularly important for the studies of nonlinear pulse propagation. In the second year of the project, we plan to purchase and use a commercially available computer-programmable pulse shaper for fast (and potentially adaptive) shaping of the laser pulses propagating through water.

Further into the project, we will expand our work into other wavelength regions, and use our two computer-controlled optical parametric amplifiers (OPerA, Coherent), pumped by our amplified femtosecond laser system (described above). Signal and idler pulses obtained from the two OPAs can be frequency-doubled or mixed with the fundamental to produce up to 20 μJ per 50 fs pulse at tunable visible wavelengths. In addition, we may use un-amplified light from the fs oscillator (at 85 MHz), for fast-repetition-rate low-peak-power experiments.

While the previous experimental work on optical precursors suffered from limited choice of laser sources, our laser system will allow great flexibility and precise control over the pulse shapes, for a broad range of well-controlled measurements.

(b) Spectrally-resolved highly-sensitive measurement at the output
We will measure the transmitted spectral intensities (and not just the total output power). We will use a state-of-the-art CCD-equipped spectrometer which will count photons at each wavelength separately. A second CCD-equipped near-infrared spectrometer will allow us to extend our overall working wavelength range. What has been used in many of the past experiments were photomultiplier tubes (PMT's). A PMT is a "bucket" detector that just counts the total number of photons and does not provide spectral resolution (unless it is combined with a spectrometer, in which case a tedious scanning

is needed). PMT's are virtually unsurpassed in sensitivity; however, a good cooled CCD can be almost as sensitive. Highly sensitive signal detection over a large dynamic range is essential for the success of the proposed project, since the intensity of light transmitted through many meters of water will vary over many orders of magnitude as a function of propagation length. Our group has a great deal of experience in performing measurements where the signal varies up to over nine decades. As an example, we reproduce here a figure[13] [Fig. 2] which shows signal varying over seven orders of magnitude. That particular set of data was obtained without spectral resolution. Currently we are capable of achieving similar or even higher sensitivity and dynamic range with spectral resolution, which is afforded by the use of a spectrometer equipped with a cooled CCD. Our measurements on pulse propagation through water will be done with spectral resolution.

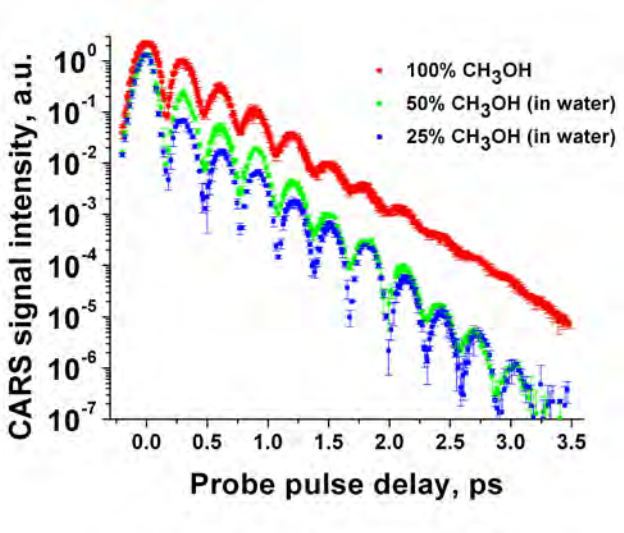

Fig. 2. An example from our previous work, where we used three ultrashort pulses at different wavelength (810 nm, 660 nm, and 575 nm) and measured femtosecond coherent anti-Stokes Raman scattering (CARS) signal (measured in methanol-water solutions) varying in magnitude over many decades[15].

At a further stage of the project we plan to measure the transmitted pulse shapes. For relatively small propagation distances (several meters) we will use a commercial autocorrelator (presently available in our lab), and a FROG apparatus (that we intend to purchase). For large propagation distances this measurement will become challenging since the output power will get very low. For the low-power measurements, we will set up a cross-correlation measurement, such as the one that has produced pictures shown in Fig. 1. Another good option for direct pulse shape measurement is an ultrafast streak camera. We have recently installed and tested such a state-of-the-art streak-camera (Hamamatsu) in our lab, and we will use it to measure the duration of pulses propagated through over 10 meters of water.

(c) Water cell design

We have designed and already constructed our own water cell. Our focus in designing the new cell was to minimize the number of mirror reflections, and to be able to increase or decrease the path lengths continuously and not just in steps of 0.5 meters.

In previous experiments, the maximum path length through the water was 6 meters. In our water cell, we will be able to run experiments at a maximum path length of 18 meters. A schematic of a simplified experimental set up is provided in Fig. 3. We currently have the equipment installed (as shown) and can have the complete setup within a short period of time.

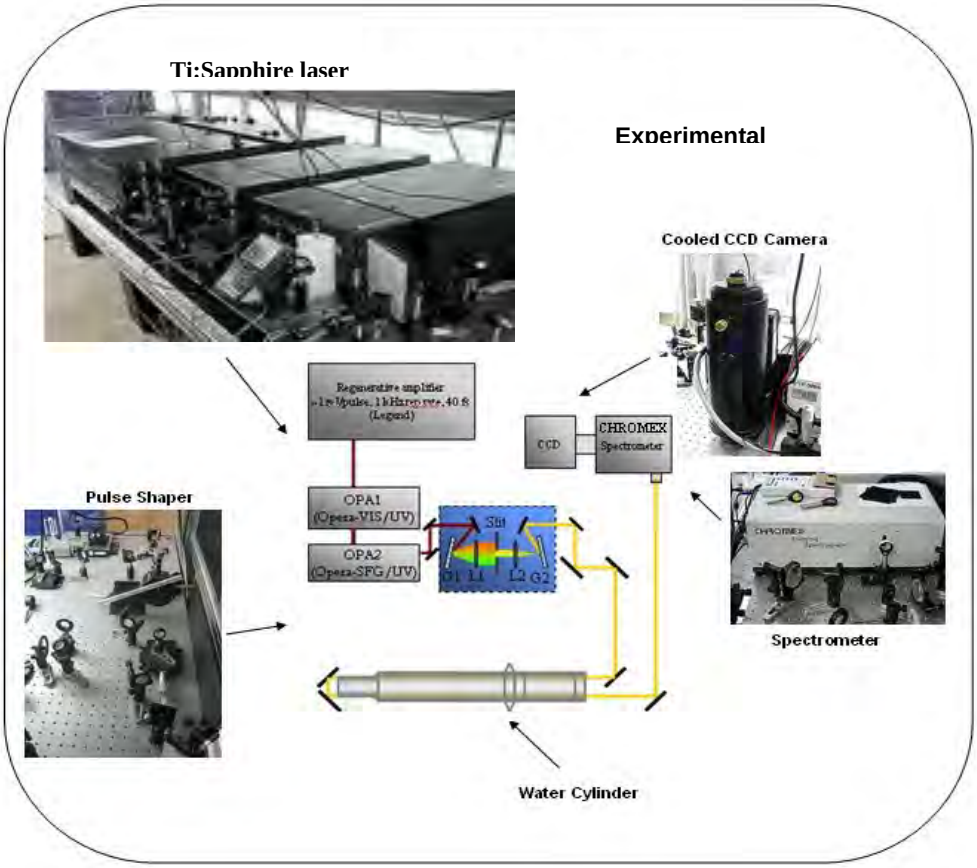

Fig. 3. Experimental setup schematics.

Theoretical:

In order to be able to model the complete pulse propagation process, we must be able to handle the actual pulse shape that will be used in our experiments along with the best data we have on the actual absorption spectra of pure water. In fact the water we will use will be prepared by the same method as in the absorption measurements done by Pope and Fry[14]. Since we have a great deal of experience with the finite difference time domain (FDTD) method, we will have to substantially rewrite parts of it to handle the actual dispersion of the refractive index of water, particularly around the overtones of the OH-stretch/scissor coupled modes. The regions around the sharp slope changes in refractive index are precisely the regions where the interesting precursors will appear. This type of approach has never been performed but is the *sine qua non* for successful modeling. The basis of our method is to take the two Maxwell curl equations in Rationalized MKSA units and assuming no currents; namely,

$$\nabla \times \vec{H} = \frac{\partial \vec{D}}{\partial t}$$
$$\nabla \times \vec{E} = -\frac{\partial \vec{B}}{\partial t} \qquad (1)$$

and then discretize them over a spatial grid. We also need the relation between the displacement current and the electric field; namely, $\vec{D} = \varepsilon(\omega)\vec{E}$ where $\varepsilon(\omega)$is the frequency dependent permittivity and is related to the refractive index *n* by $n^2 = \varepsilon(\omega)$. In the many papers we have published using the FDTD method to solve scattering from irregularly shaped particles (see references 15-26), we have worked only at a single frequency since the incoming radiation was assumed to be monochromatic. Now when using short pulses, there are a plethora of frequencies involved and so then we must be able to use the actual dispersion relation for the medium involved. In the case of water in the near IR (our pulses are centered around 800 nm) we will use the actual measured absorption data of either Kuo or Segelstein in order to apply the FDTD method to this problem, we need to convert the frequency dependent permittivity to a time dependent one. In particular, the temporal characteristic of the water permittivity should be modeled around the overtones of the OH-stretch/scissor coupled modes. These are the regions where the sharp changes in the slope of the absorption coefficient, denoted by α, occur. Another very interesting aspect of water occurs when it is deuterated to form either HDO or D_2O. These forms will have radically different absorption spectra and we can further test our analyses by studying these different forms of water.

If we denote the refractive index of water by $n=n_R-in_I$ and the permittivity by $\varepsilon=\varepsilon_R-i\varepsilon_I$, where the subscripts R and I denote real and imaginary parts respectively, then it is easy to show that $\varepsilon_R = n_R^2 - n_I^2$ and $\varepsilon_I = 2n_R n_I$. The absorption coefficient α is related to n_I by $\alpha = 4\pi n_I / \lambda_{vac}$ where λ_{vac} denotes the wavelength of the radiation in vacuo.

In order to get the time dependence of water permittivity, we must Fourier transform $\varepsilon(\omega)$. Actually it is more physical to Fourier transform $\varepsilon(\omega)$-1 and to obtain the susceptibility kernel $G(\tau)$ defined by (see Jackson[27]):

$$G(\tau) = \frac{1}{2\pi}\int_{-\infty}^{\infty}[\varepsilon(\omega)-1]e^{-i\omega\tau}d\omega \qquad (3)$$

then

$$\vec{D}(\vec{x},t) = \vec{E}(\vec{x},t) + \int_{-\infty}^{\infty} G(\tau)\vec{E}(\vec{x},t-\tau)d\tau$$
$$= \vec{E}(\vec{x},t) + \sum_{\tau=0}^{t} G(\tau)\vec{E}(\vec{x},t-\tau)\Delta\tau \qquad (4)$$

Thus we see that the displacement current at a certain instant in time, depends on the electric field from earlier times. It should also be noted that if ε is independent of frequency, then Eq. (4) reduces to $\vec{D} = \varepsilon\vec{E}$which is what our present FDTD code now uses.

Once we have established the fact that the FDTD method is a viable one for solving this problem, then we would like to next explore the possibility of using the pseudospectral time domain (PSTD) method which we feel will run much faster than the conventional FDTD method. We have tested this method on some canonical scattering problems and found substantial reductions in computation time when compared to the conventional FDTD method. With this method we will actually be able to monitor the complete temporal evolution of the pulse shape.

WORK COMPLETED

a) We have demonstrated energy coupling between filament-forming pulsed laser beams in a bulk medium (methanol) [1]. Furthermore, we have identified a previously unreported phenomenon in which the energy transfer between beams oscillates on a time scale equal to the temporal period of the incident laser radiation (2.6 fs). This demonstrates that filament propagation can be finely controlled and has applications in atmospheric and underwater remote sensing.

b) We have developed several techniques to image propagating laser filaments with the goal of studying their interactions with the propagating medium. Our goal is to use such methods in the study of the acoustic signal generated by propagating filaments.

c) We have demonstrated the power and efficacy of filamentation-induced breakdown spectroscopy (FIBS) in a proof-of-principle experiment. Our results indicate that a myriad of different materials may be quickly and accurately identified from a great distance. We demonstrate that FIBS is a new tool that has powerful applications in atmospheric and underwater remote sensing.

RESULTS

We first present the results from our experiment with intersecting filament-forming pulsed laser beams in liquid methanol [1]. An experimental schematic is presented in Fig. 1.

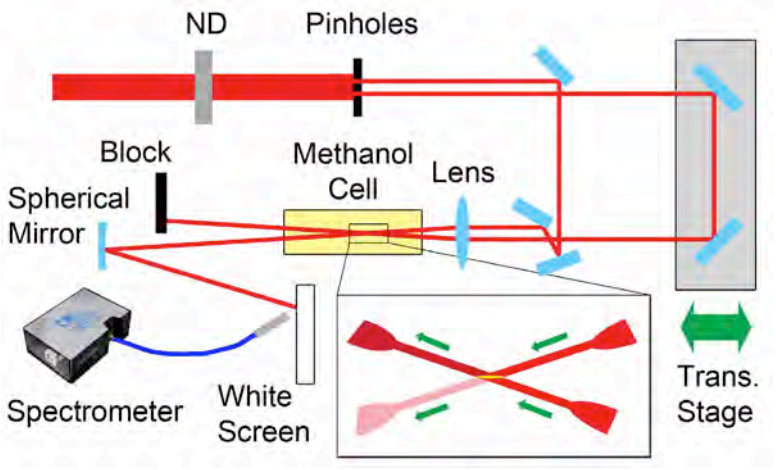

Fig. 1 Experimental schematic for measuring behavior of two intersecting filaments in liquid methanol.

Two filaments were intersected within liquid methanol and their output powers were measured. We observed energy transfer similar to that of previous experiments involving two intersecting laser pulses in air [2, 3] in which energy transfer varies on a time scale of 100 fs relative pulse delay. We also observed a dependence on pulse energy which is consistent with results in [3]. Such behavior is depicted in Fig. 2.

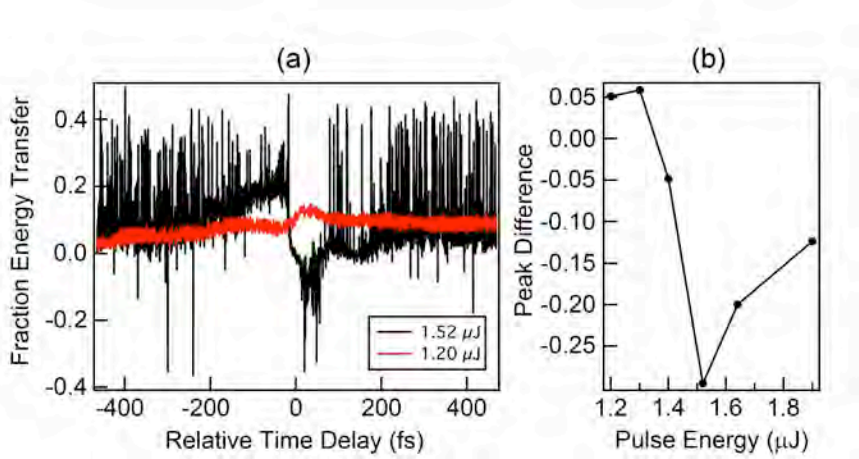

Fig. 2. (a) Energy transfer curves between two filaments for pulse energies 1.52 μJ and 1.20 μJ. Note that energy transfer changes direction. (b) The difference between peaks in the energy transfer curve for pulses with varying energy. Such behavior is consistent with that in [3].

In addition to observing behavior consistent with intersecting filaments in air, we have identified a phenomenon in which energy transfer occurs between the two beams on the order of the temporal period of the incident radiation (2.6 fs). This is shown in Fig. 3.

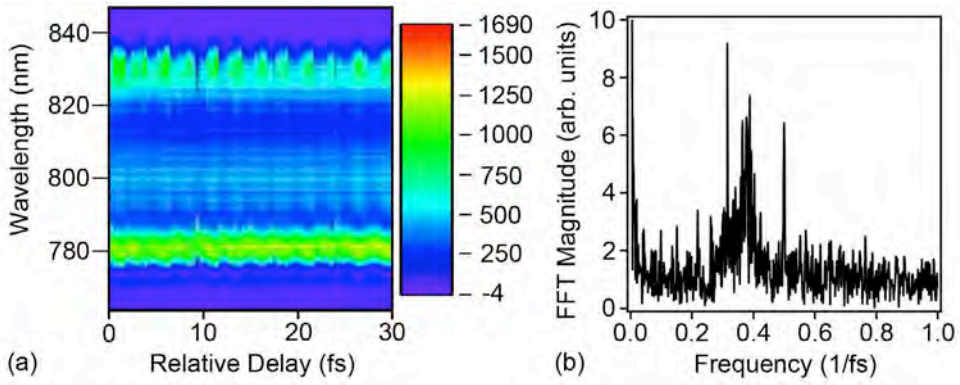

Fig. 3. (a) Raw spectral measurement showing pulse spectrum as a function of relative pulse delay. (b) FFT of (a) showing that the frequency of the oscillations is equal to that of the laser pulse central frequency.

We show in Fig. 4 that the energy in the beams oscillate out of phase with one another.

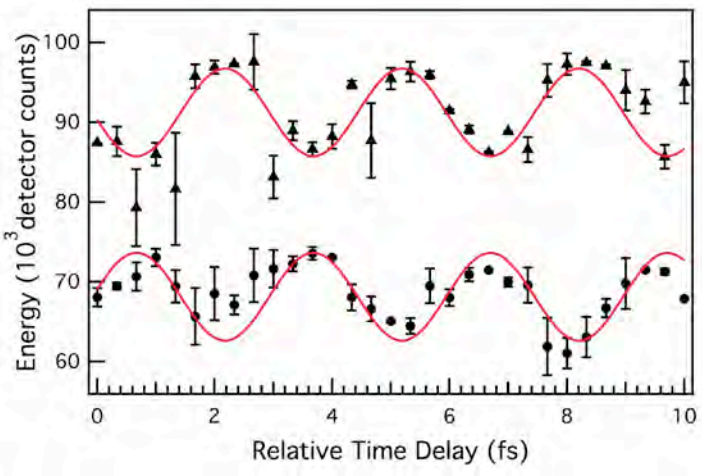

Fig. 4. Energy in each beam as a function of relative pulse delay. The upper curve has been displaced by 20 *10^3 detector counts.

In conclusion, we have demonstrated that intersecting filaments in a bulk medium exhibit behavior similar to that of intersecting filaments in air. Also, we have reported a previously undocumented phenomenon in which the energy in each beam oscillates out of phase with the other at a frequency equal to that of the laser pulse central frequency. Such results indicate that filament propagation can be finely controlled and that they might be used for sophisticated applications in atmospheric and underwater remote sensing.

We have also developed several techniques for the imaging of intense laser pulses in air and bulk media such as water and methanol. These techniques include shadowgraphy and Schlieren photography variants as well as polarization-selective imaging from induced birefringence. Shown in Fig. 5 is an experimental setup for imaging with a Schlieren-type photographic method. Fig. 6 shows an example of an intense femtosecond laser pulse propagating through air imaged with this setup.

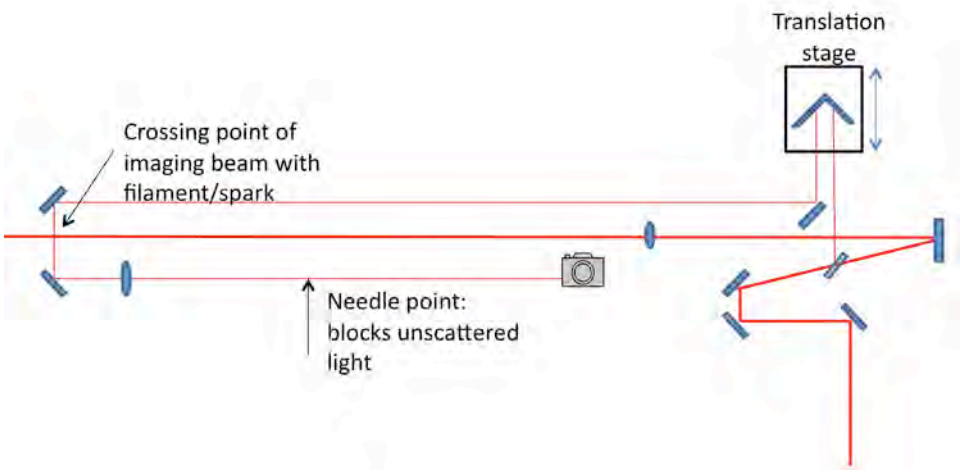

Fig. 5. Experimental setup for imaging intense femtosecond laser pulses with a Schlieren-type photographic method.

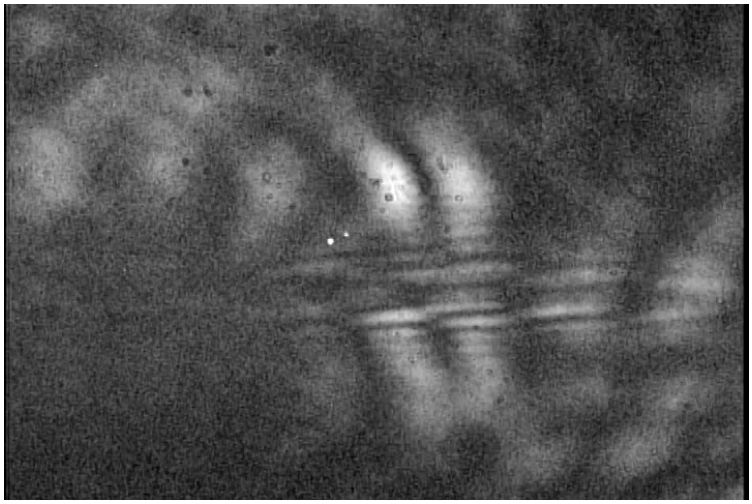

Fig. 6. Photograph of an intense femtosecond laser pulse propagating through air imaged with a Schlieren-type photographic method.

These imaging techniques are pursued with the goal of studying the acoustic signal from a propagating femtosecond laser filament in water. Understanding the acoustic signal from filaments may have applications in underwater communications and remote sensing.

Finally, we present results from a proof-of-principle experiment using filament-induced breakdown spectroscopy (FIBS) to distinguish between different soil, rock, and biological samples. An experimental schematic is shown in Fig. 7.

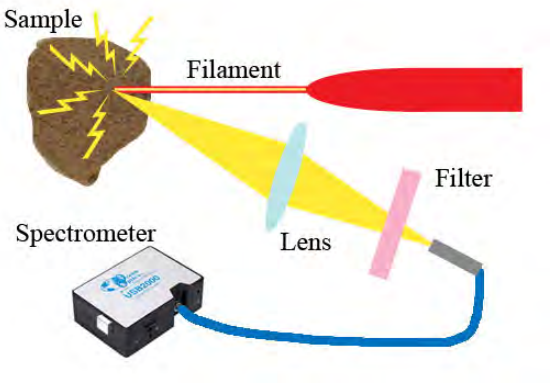

Fig. 7. Experimental setup for measuring spectra from samples ablated with a femtosecond laser filament.

We analyzed spectra from both wet and dry samples of soil, sand, and clay. Results are shown in Fig. 8.

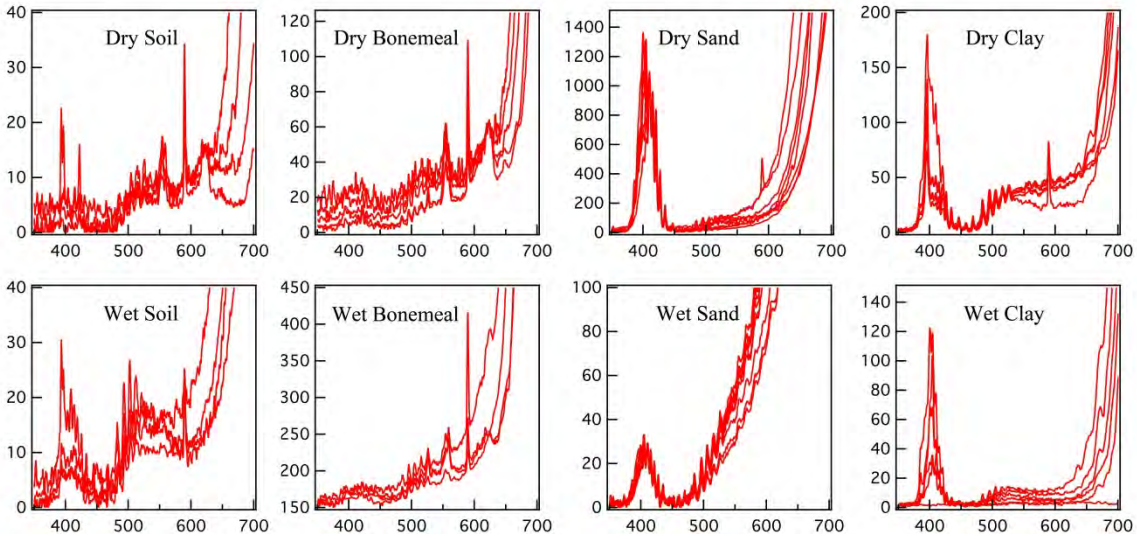

Fig. 8. Spectra from ablated samples of dry and wet soil, bonemeal, sand, and clay. Notice that each sample may be distinguished from the others, and that wet samples may be distinguished from their dry counterparts.

The results indicate that we are able to distinguish between soil, bonemeal, sand, and clay samples. We are also able to distinguish between dry and wet samples. The power and versatility of the technique are demonstrated in the variety of samples able to be distinguished. Additional samples of metals, rocks, and biological materials are shown in Fig. 9.

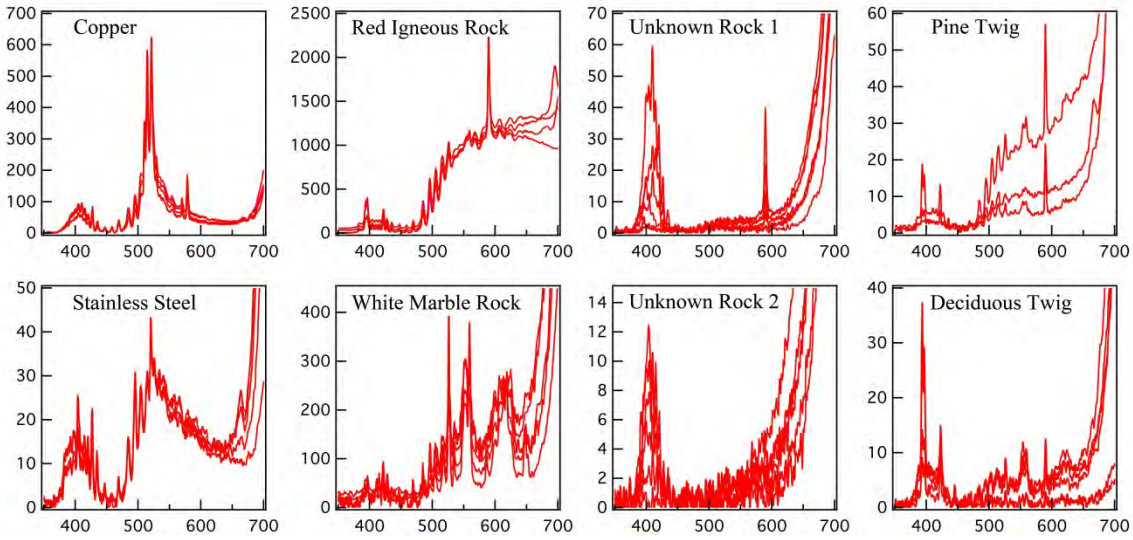

Fig. 9. Spectra from ablated samples of copper, stainless steel, red igneous rock, white marble rock, two unknown rocks, and pine and deciduous twigs.

These spectra were collected with an non-gated measurement technique in which the signal from the ablated samples was integrated over 50 to 100 ms. We fully expect to improve upon these results with implementation of a time-gated measurement technique in which the signal from ablated signals may be selectively integrated over time scales of nanoseconds or microseconds.

Our results show that FIBS is a powerful new technology which has yet to be fully explored and implemented. We plan to complete additional experiments which will demonstrate the versatility and applicability of the technique in remote sensing applications.

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